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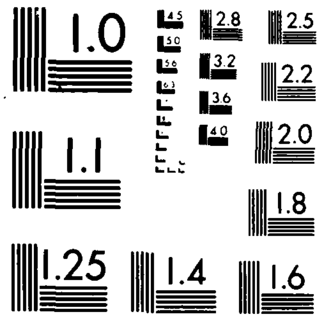

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# EQUATION OF STATE AND SOUND VELOCITIES FROM ISOTROPIC CONTINUUM MECHANICS


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OCTOBER 1986

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We use the methods of finite elasticity in continuum mechanics of homogeneous isotropic materials to obtain the pressure dependence of the equation of state and the shear and longitudinal velocity to fifth order elastic constants. The resulting expressions are implicit in terms of the pressure and explicit in terms of the strain. The use of a symbolic program allows us to eliminate the strain parameter. Even though the expressions are extremely lengthy,

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20. ABSTRACT (CONT'D)

approximations for each order of elastic constants can be easily obtained using Taylor's expansion to any degree desired.

TABLE OF CONTENTS



LIST OF ILLUSTRATIONS

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## INTRODUCTION

The equation of state (EOS) is central to the determination of physical properties at high pressure. In the pressure range where solid high pressure systems have been used, the lack of a primary or direct means to measure the pressure has compounded the difficulty of obtaining the high pressure EOS of a solid. The solution came together in the Decker equation of state for sodium chloride (NaCl) (ref 1). Decker calculated the EOS on the basis of the Mie-Gruneisen equation. The Decker equation had multiple virtues; it produced agreement with the best pressures data (thus tending to validate the pressure scale as well), had no adjustable parameters, and dealt with NaCl, a very popular high pressure material. It was used as an encapsulant for obtaining "hydrostatic" pressure in solid systems and in experiments in which x-rays were used.

After having made sound velocity measurements in NaCl to 270 kbar (ref 2), we addressed the problem of the EOS (ref 3) following Murnaghan's (ref 4) developments in continuum mechanics using finite deformation. We extended the calculations to fourth order elastic constants for the equation of state and also, on the basis of the same formulation, obtained expressions for the longitudinal and shear velocities as a function of the hydrostatic strain

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[1] D. L. Decker, W. A. Barrett, L. Merrill, H. T. Hall, and J. D. Barnet, J. Phys. Chem., Vol. 1, No. 3, 1972, p. 773.

[2] J. Frankel, J. F. Rich, and C. G. Homan, J. Geoph. Res., Vol. 81, 1976, p. 6357.

[3] J. Frankel, M. A. Hussain, and R. D. Scanlon, J. Phys. Chem. Solids, Vol. 40, 1979, p. 67.

[4] F. D. Murnaghan, Finite Deformations of an Elastic Solid, Dover Publications, Inc., New York, 1967.

(also to fourth order elastic constants). Finite elasticity is essential where the elastic deformations are so large that Hooke's law representing the deformation by means of second order ($\lambda$ and $\mu$) elastic constants is an oversimplification. Figure 1 shows a representation of the EOS for NaCl. It can be seen that the (Hookean) second order equation of state is inadequate and that the EOS using fourth order elastic constants approximates the Decker equation better.

The third order fit is identical to the Murnaghan (ref 4) equation. The elastic constants used in Figure 1 to calculate the EOS to 270 kbar were obtained from ambient pressure data and from the measurements of Voronov and Grigorev (ref 5) to 80 kbar. In Reference 3, however, we see that these same elastic constants do not fit the velocities measured experimentally beyond 80 kbar. The velocities present a more stringent test of the theory than the EOS.

The remainder of this report will be devoted to a continuation of the work commenced in Reference 3, an extension of the theory to fifth order elastic constants, and an attempt to express the velocities in terms of pressure. Expressions for velocities in terms of pressure rather than strain, facilitate comparison to simpler expressions in the literature, which were derived for lower order elastic constants, and also invite comparison with experimental data.

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[3] J. Frankel, M. A. Hussain, and R. D. Scanlon, J. Phys. Chem. Solids, Vol. 40, 1979, p. 67.

[4] F. D. Murnaghan, Finite Deformations of an Elastic Solid, Dover Publications, Inc., New York, 1967.

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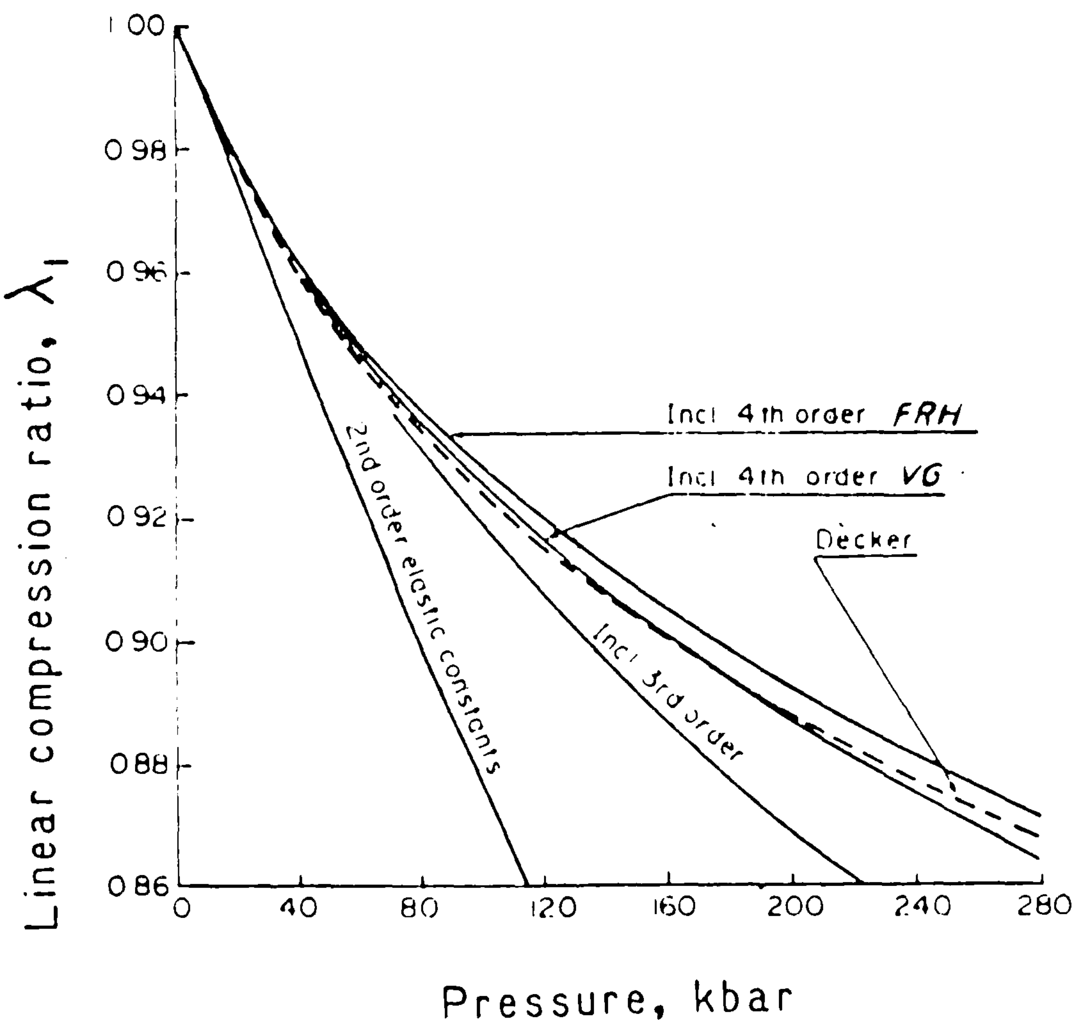


Figure 1. The representation of the EOS for NaCl with inclusion of second, third, and fourth order elastic constants. The best fit to 280 kbar results when using fourth order elastic constants obtained by Voronov and Grigorev (ref 5) data for 80 kbar (ref 3). $\lambda_1$ is defined as the ratio of the dimension at pressure to the original dimension.

In the next section, explicit expressions are derived in terms of the strain parameter $\lambda_1$. In the following section, we obtain the direct relations between velocity and pressure by eliminating the strain parameter via the EOS. The expressions become unmanageable and we carry out a Taylor expansion around the zero pressure state. Without the use of the symbolic manipulation algorithm (M'.CSYMA) (ref 6) many of these expressions could not have been obtained easily.

## OUTLINE OF ANALYSIS AND ALGORITHM

For completeness we briefly outline the algorithm for obtaining the velocities and the equation of state. The complete analysis was given in Reference 3. We apply a uniform hydrostatic deformation to the unconstrained body with coordinates $B_o(x_1,y_1,z_1)$ which become $B(x,y,z)$ in the deformed state. The strain parameter $\lambda_1$ is given by

$$\lambda_1 = \frac{x}{x_1} = \frac{y}{y_1} = \frac{z}{z_1} \tag{1}$$

The metric tensor $g_{ij}, g^{ij}$ of the undeformed body and $G_{ij}$, $G^{ij}$ of the deformed body are given by

$$g_{ij} = \begin{bmatrix} \lambda_1^{-2} & 0 & 0 \\ 0 & \lambda_1^{-2} & 0 \\ 0 & 0 & \lambda_1^{-2} \end{bmatrix}, \quad g^{ij} = \begin{bmatrix} \lambda_1^{2} & 0 & 0 \\ 0 & \lambda_1^{2} & 0 \\ 0 & 0 & \lambda_1^{2} \end{bmatrix} \tag{2}$$

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[3]J. Frankel, M. A. Hussain, and R. D. Scanlon, J. Phys. Chem. Solids, Vol. 40, 1979, p. 67.

[6]"MACSYMA" Reference Manual, The Mathlab Group, Laboratory for Computer Science, MIT, MA, Version Ten, 1983.

$$G_{ij} = G^{ij} = \delta_{ij} \tag{3}$$

The strain invariants are

$$I_1 = 3\lambda_1^{\,2} \quad , \quad I_2 = 3\lambda_1^{\,4} \quad , \quad I_3 = \lambda_1^{\,6} \tag{4}$$

The relation of the above strain invariants to those given by Murnaghan (ref 4) is given by the following expressions, where we use superscript c to denote the definition of Murnaghan:

$$I_1^{\,c} = \frac{1}{2}\,(I_1-3) \quad , \quad I_2^{\,c} = \frac{1}{4}\,[(I_2-3) - 2(I_1-3)]$$

$$I_3^{\,c} = \frac{1}{8}\,[(I_3-1) + (I_1-3) - (I_2-3)] \tag{5}$$

and the strain tensor becomes

$$\gamma_{ij} = \frac{1}{2}\,(G_{ij}-g_{ij}) = \frac{1}{2}\,\delta_{ij}(1 - \lambda_1^{\,-2}) \tag{6}$$

Expressing the strain energy density W in terms of $I_1$, $I_2$, and $I_3$, we obtain from the principle of least action the components of the stress tensor

$$\tau^{11} = \phi\lambda_1^{\,2} + 2\psi\lambda_1^{\,4} + P$$

$$\tau^{22} = \tau^{33} = \tau^{11}$$

$$\tau^{12} = \tau^{13} = \tau^{23} = 0 \tag{7}$$

$$\phi = \frac{2}{\sqrt{I_3}}\,\frac{\partial W}{\partial I_1} \qquad \psi = \frac{2}{\sqrt{I_3}}\,\frac{\partial W}{\partial I_2} \qquad P = 2\sqrt{I_3}\,\frac{\partial W}{\partial I_3} \tag{8}$$

From the perturbation analysis given in Reference 1, we have the following expressions for the velocities and the equation of state (in terms of the strain parameter $\lambda_1$ and derivatives of the strain energy W):

---

[1]D. L. Decker, W. A. Barrett, L. Merrill, H. T. Hall, and J. D. Barnet, J. Phys. Chem., Vol. 1, No. 3, 1972, p. 773.

$$v_p = \left(\frac{c_{11}+\tau^{11}}{\rho}\right)^{1/2} \qquad \text{longitudinal velocity}$$

$$v_s = \left(\frac{c_{11}-c_{12}+2\tau^{11}}{2\rho}\right)^{1/2} \qquad \text{shear velocity}$$

$$-p = \phi\lambda_1^2 + 2\psi\lambda_1^4 + P \qquad \text{equation of state}$$

where p is the hydrostatic pressure and P is defined in Eq. (8).

$$c_{11} = -\tau^{11} + 2A\lambda_1^4 + 8B\lambda_1^8 + 2C\lambda_1^{12} +$$
$$8D\lambda_1^{10} + 4E\lambda_1^8 + 8F\lambda_1^6$$
$$c_{12} = -\lambda_1^2\phi + P + 2A\lambda_1^4 + 8B\lambda_1^2 + 2C\lambda_1^{12}$$
$$+ 8D\lambda_1^{10} + 4E\lambda_1^8 + 8F\lambda_1^6$$

$$c_{44} = \frac{1}{2}(c_{11}-c_{12}) \tag{9}$$

and

$$A = \frac{2}{\sqrt{I_3}}\frac{\partial^2 W}{\partial I_1^2}, \quad B = \frac{2}{\sqrt{I_3}}\frac{\partial^2 W}{\partial I_2^2}$$

$$C = \frac{2}{\sqrt{I_3}}\frac{\partial^2 W}{\partial I_3^2}, \quad D = \frac{2}{\sqrt{I_3}}\frac{\partial^2 W}{\partial I_2 \partial I_3}$$

$$E = \frac{2}{\sqrt{I_3}}\frac{\partial^2 W}{\partial I_3 \partial I_1}, \quad F = \frac{2}{\sqrt{I_3}}\frac{\partial^2 W}{\partial I_1 \partial I_2} \tag{10a}$$

and from Murnaghan (ref 4), the expressions for the density become

$$\rho = \rho_o[1 + 2I_1{}^c + 4I_2{}^c + 8I_3{}^c]^{-1/2} = \rho_o(I_3)^{-1/2} \tag{10b}$$

---

[4]F. D. Murnaghan, Finite Deformations of an Elastic Solid, Dover Publications, Inc., New York, 1967.

## DERIVATION FOR HIGHER ORDER ELASTIC CONSTANTS IN INCREASING ORDER

### Second Order Elastic Constants

The strain energy in terms of second order elastic constants becomes

$$W = \frac{1}{2}(\lambda+2\mu)(I_1{}^c)^2 - 2\mu I_2{}^c \tag{11}$$

where $\lambda$ and $\mu$ are the classical second order elastic constants. Using Eqs. (9) and (10), we have

$$\rho v_p{}^2 = \lambda\left(\frac{5\lambda_1}{2} - \frac{3}{2\lambda_1}\right) + \mu\left(3\lambda_1 - \frac{1}{\lambda_1}\right) \tag{12}$$

$$\rho v_s{}^2 = \frac{3\lambda}{2}\left(\lambda_1 - \frac{1}{\lambda_1}\right) + \mu\left(2\lambda_1 - \frac{1}{\lambda_1}\right) \tag{13}$$

$$-P = \frac{1}{2}(3\lambda+2\mu)\lambda_1{}^{-1}(\lambda_1{}^2-1) \tag{14}$$

$$\rho = \rho_o/\lambda_1{}^3$$

Eliminating the strain parameter between Eqs. (12) and (14), we obtain a quadratic expression for $v_p{}^2$; a solution is given by

$$\begin{aligned}\rho_o v_p{}^2 = \{&81\lambda^5 + 378\mu\lambda^4 - [9\lambda^2 + 12\mu\lambda + p^2 + 4\lambda^2]^{1/2}[63p\lambda^3 + 174\mu p\lambda^2 + \\ &(20p^3 + 148\mu^2 p)\lambda + 24\mu p^3 + 40\mu^3 p] + (153p^2 + 648\mu^2)\lambda^3 + \\ &(402\mu p^2 + 528\mu^3)\lambda^2 + (20p^4 + 332\mu^2 p^2 + 208\mu^4)\lambda + \\ &(24\mu p^4 + 88\mu^3 p^2 + 32\mu^5)\}/(81\lambda^4 + 216\mu\lambda^3 + 216\mu^2\lambda^2 + 96\mu^3\lambda + 16\mu^4)\end{aligned} \tag{15}$$

Equation (15), though exact, is more cumbersome than a Taylor expansion of itself up to $p^3$. This becomes

$$\rho_o v_p^{\,2} = (\lambda+2\mu) - \frac{(7\lambda+10\mu)p}{3\lambda+2\mu} + \frac{(17\lambda+22\mu)p^2}{(3\lambda+2\mu)^2} - \frac{(47\lambda+58\mu)p^3}{2(3\lambda+2\mu)^3} \tag{16}$$

We followed a similar procedure for the shear velocity. The Taylor expansion for the shear velocity yields

$$\rho v_s^{\,2} = \mu - \frac{(3\lambda+6\mu)p}{3\lambda+2\mu} + \frac{(9\lambda+14\mu)p^2}{(3\lambda+2\mu)^2} - \frac{(27\lambda+38\mu)p^3}{2(3\lambda+2\mu)^3} \tag{17}$$

Equation (16) does not agree with that given by Birch (ref 7). The discrepancy may be due to an incorrect expression of Eq. (10b) by Birch. (See corresponding expression in Reference 4, p. 37).

Third Order Elastic Constants

In the following (Eqs. (18) through (21)), we will only add the additional terms due to the third order analysis. The strain energy density for the third order elastic constants becomes

$$W = \frac{(\ell+2m)}{3}(I_1^{\,c})^3 - 2mI_1^{\,c}I_2^{\,c} + nI_3^{\,c} \tag{18}$$

As shown in Reference 3, for our problem the three Murnaghan third order elastic constants $\ell$, m, and n only appear as two third order elastic constants

$$\left.\begin{aligned} \alpha &= (9\ell + n) \\ \beta &= (3\ell + 2m) \end{aligned}\right\} \text{ Third Order}$$

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[3]J. Frankel, M. A. Hussain, and R. D. Scanlon, J. Phys. Chem. Solids, Vol. 40, 1979, p. 67.
[4]F. D. Murnaghan, Finite Deformation of an Elastic Solid, Dover Publications, Inc., New York, 1967.
[7]F. Birch, J. Appl. Phys., Vol. 9, 1938, p. 279.

The velocities and the EOS have the following terms in addition to those found from the second order terms:

$$\text{for } \rho v_p{}^2 = \lambda_1{}^3(\frac{1}{4}\alpha+\beta) + \lambda_1(-\frac{1}{2}\alpha-\beta) + \lambda_1{}^{-1}(\frac{1}{4}\alpha) \tag{19}$$

$$\text{for } \rho v_s{}^2 = \lambda_1{}^3(\frac{3}{4}\beta) + \lambda_1(-\frac{1}{4}\alpha - \frac{3}{4}\beta) + \lambda_1{}^{-1}(\frac{1}{4}\alpha) \tag{20}$$

$$\text{for } -p = \frac{\alpha}{4\lambda_1}(\lambda_1{}^2-1)^2 \tag{21}$$

As before, we eliminate $\lambda_1$ for $\rho v_p{}^2$ and $\rho v_s{}^2$ which include second order terms and the third order terms between Eqs. (19) and (21) and Eqs. (20) and (21), respectively. The resulting expressions are very complex algebraically; as before, a more useful form is obtained from a Taylor expression of the exact function for velocity. The resulting expressions for the velocities in terms of second and third order elastic constants are

$$\rho_o v_p{}^2 = \lambda + 2\mu - \frac{(7\lambda+10\mu+2\beta)p}{3\lambda+2\mu} + \frac{(17\lambda + 22\mu + 10\beta + \alpha)p^2}{(3\lambda+2\mu)^2} - \frac{(47\lambda+58\mu+50\beta+8\alpha)p^3}{2(3\lambda+2\mu)^3} + \ldots \tag{22}$$

$$\rho_o v_s{}^2 = \mu - \frac{(6\lambda+12\mu+3\beta-\alpha)p}{6\lambda+4\mu} + \frac{(18\lambda+28\mu+15\beta-3\alpha)p^2}{2(3\lambda+2\mu)^2} - \frac{(54\lambda+76\mu+75\beta-9\alpha)p^3}{4(3\lambda+2\mu)^3} + \ldots \tag{23}$$

Fourth Order Elastic Constants

$$W = 16q(I_1^c)^4 + 16r(I_1^c)^2(I_2^c) + 16s(I_1^c)(I_3^c) + 16t(I_2^c)^2 \tag{24}$$

Two fourth order elastic constants appear as follows:

$$\left.\begin{aligned} \gamma &= (27q + 9r + s + 3t) \\ \delta &= (81q + 24r + 2s + 7t) \end{aligned}\right\} \text{Fourth Order} \tag{25}$$

Velocities and pressure are given with additional terms

$$v_p^2\rho = \lambda_1^5(8\delta) + \lambda_1^3(-8\gamma-16\delta) + \lambda_1(16\gamma+8\delta) + \lambda_1^{-1}(-8\gamma) \tag{26}$$

$$v_s^2\rho = \lambda_1^5(6\delta-10\gamma) + \lambda_1^3(-12\delta+12\gamma) + \lambda_1(6\gamma+6\delta) + \lambda_1^{-1}(-8\gamma) \tag{27}$$

$$-p = \frac{8\gamma}{\lambda_1}(\lambda_1^2-1)^3 \tag{28}$$

The Taylor expansion, after an elimination procedure, gives the velocities as functions of pressure

$$\rho_o v_p^2 = (\lambda+2\mu) - \frac{(7\lambda+10\mu+2\beta)p}{3\lambda + 2\mu} + \frac{(17\lambda+22\mu-32\gamma+32\delta+10\beta+\alpha)p^2}{(3\lambda+2\mu)^2} - \frac{(47\lambda+58\mu-256\gamma+384\delta+50\beta+8\alpha)p^3}{2(3\lambda+2\mu)^3} + \ldots \tag{29}$$

$$\rho_o v_s^2 = \mu - \frac{(6\lambda+12\mu+3\beta-\alpha)p}{6\lambda + 4\mu} + \frac{(18\lambda+28\mu-144\gamma+48\delta+15\beta-3\alpha)p^2}{2(3\lambda+2\mu)^2} - \frac{(54\lambda+76\mu-1472\gamma+576\delta+75\beta-9\alpha)p^3}{4(3\lambda+2\mu)^3} + \ldots \tag{30}$$

## Fifth Order Elastic Constants

$$W = 32a(I_1^c)^5 + 32b(I_1^c)^3(I_2^c) + 32c(I_1^c)^2(I_3^c)$$
$$+ 32d(I_1^c)(I_2^c)^2 + 32e(I_2^c)(I_3^c) \qquad (31)$$

Two fifth order elastic constants appear as follows:

$$\left.\begin{aligned} \omega &= 108a + 27b + c + 6d \\ \varepsilon &= 81a + 27b + 3c + 9d + e \end{aligned}\right\} \text{Fifth Order} \qquad (32)$$

$$\rho v_p^2 = \frac{1}{\lambda_1}(\lambda_1^2-1)^3[(8\omega+26\varepsilon)\lambda_1^2-10\varepsilon] \qquad (33)$$

$$\rho v_s^2 = 3\lambda_1(\lambda_1^2-1)^3(2\omega-\varepsilon) \qquad (34)$$

$$-p = \frac{\varepsilon}{\lambda_1}10(\lambda_1^2-1)^4 \qquad (35)$$

Finally, we carry out Taylor's expansion after the elimination process. This time, however, the velocities are not affected up to the second order in p, i.e., Eqs. (29) and (30) remain valid with $p^3$ term given below

$p^3$ term in $\rho_o v_p^2$

$$-(47\lambda + 128\omega + 58\mu - 256\gamma + 256\varepsilon + 384\delta + 50\beta + 8\alpha)p^3/2(3\lambda+2\mu)^3 \qquad (36)$$

and

$p^3$ term in $\rho_o v_s^2$

$$-(54\lambda + 192\omega + 76\mu - 1472\gamma - 96\varepsilon + 576\delta + 75\beta - 9\alpha)p^3/4(3\lambda+2\mu)^3 \qquad (37)$$

## DISCUSSION AND CONCLUSION

Figures 2 and 3 give results in which the exact solution for $v_p$ and $v_s$ to fourth order elastic constants for NaCl are compared with Eqs. (29) and (30), respectively. In both cases, the expressions are essentially identical to 30 kbar. Velocity data under hydrostatic conditions can be obtained to good accuracy up to 25 kbar. For materials with smaller compressibility than sodium chloride, plotting the measured velocities, Eqs. (29) and (30) to approximately 25 kbar can yield the elastic constants which would give us the equation of state when no phase changes occur. More compressible materials would require fifth order elastic constants.

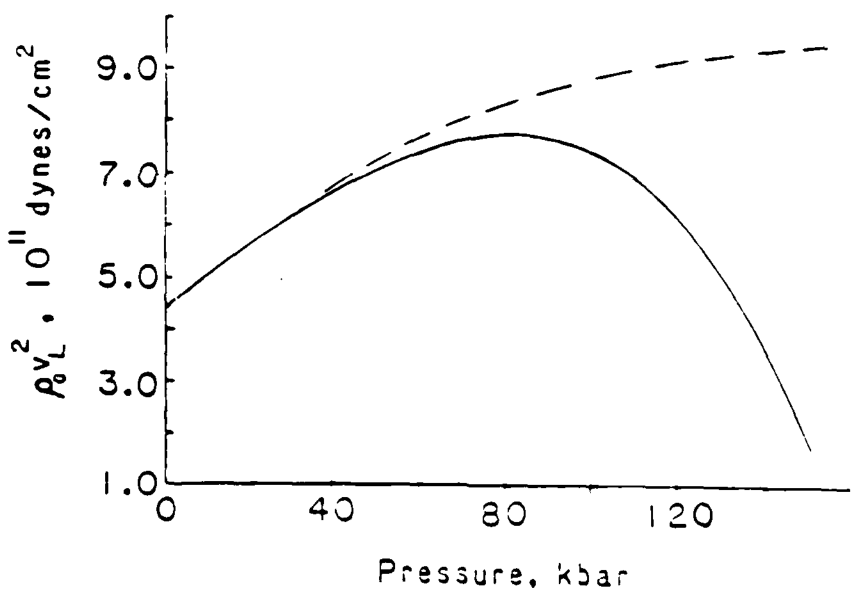


Figure 2. $\rho_o v_p^2$ versus pressure. Exact solution (---) and Taylor expansion (____) (Eq. (29)) using the elastic constants for NaCl given in Reference 3.

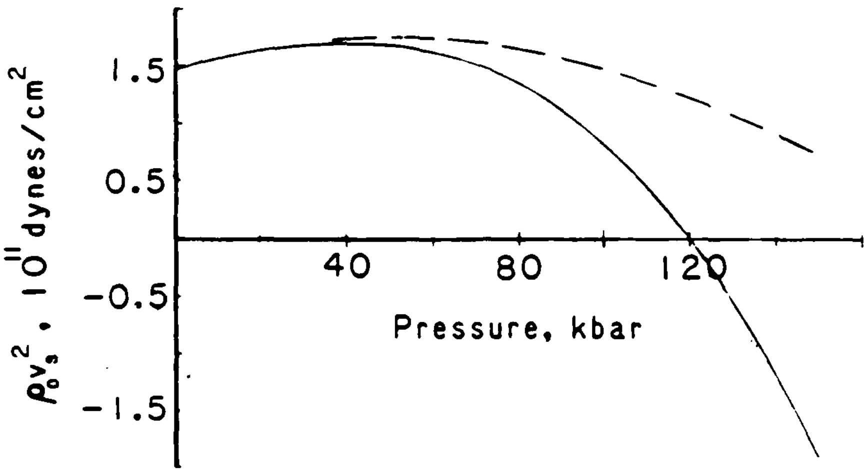


Figure 3. $\rho_o v_s{}^2$ versus pressure. Exact solution (---) and Taylor expansion (___) (Eq. (30)) using the elastic constants for NaCl given in Reference 3.

There is an exact solution on the basis of a classical elastic theory for velocity as a function of pressure. Just as x-rays were used to calculate pressure via lattice spacings of sodium chloride, it is not hard to visualize using velocities to do the same thing.

Brillouin scattering in a diamond cell could be used and data would have to be obtained from two materials, not necessarily embedded in each other, but at the same pressure in the cell.

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